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This paper presents frequency doubling results by Cerenkov radiation from stable poled polymer slab waveguides. For a 1064 nm Nd-YAG laser, the harmonic wavelength is away from the strong absorption region of the nonlinear optical polymer used. Green light was observed even when the laser was operating at a continuous wave 830 nm diode laser and a polymer with a larger nonlinear coefficient. The waveguide conversion efficiency for doubling of the diode laser was estimated to be 0.03 %/W with a total efficiency of 2.8 x 10-5 %/W.

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by

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EFFICIENT CERENKOV SECOND-HARMONIC GENERATION IN CROSSLINKED POLED POLYMER WAVEGUIDES

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This paper presents frequency doubling results by Cerenkov radiation from stable poled polymer slab waveguides. For a 1064-nm Nd-YAG laser, the harmonic wavelength is away from the strong absorption region of the nonlinear optical polymer used. Green light was observed even when the laser was operating at a continuous wave mode. Further, blue light was obtained using a continuous wave 830-nm diode laser and a polymer with a larger nonlinear coefficient. The waveguide conversion efficiency for doubling of the diode laser was estimated to be 0.03 %/W with a total efficiency of 2.8×10^{-5} %/W.

1. Introduction

Second-harmonic generation (SHG) of laser light in poled polymer waveguide is a promising way to obtain inexpensive, efficient, and compact short wavelength coherent sources. Among various approaches [1,2] to realize such SHG devices, Cerenkov radiation [3] is attractive since the phase-matching condition is not stringent and the nonlinear optical (NLO) polymer can be lossy at the second-harmonic (SH) wavelength. Sugihara *et al.* have recently reported Cerenkov SHG from a copolymer of methyl methacrylate and Disperse Red 1 substituted methylacrylate.[4] However, such materials exhibit slow relaxation of nonlinearity and have a smaller SHG coefficient. Moreover, effective interaction length cannot be long if the substrate transmitting Cerenkov radiation is thin and the absorption by the NLO polymer is strong at the SH frequency 2ω.

For poled polymers, it is important to maintain the alignment of the NLO moieties against thermal relaxation.[5,6] We have recently utilized a photocrosslinking technique for such purpose, where crosslinking of the aligned molecules is performed by irradiating the polymer with UV light during the late stage of poling.[7-10] These materials can be directly patterned by shining light through a mask for making channel waveguides and other integrated optical devices.[11] Thus, this type of materials provide both ease of processing and stable nonlinear response.

In this paper, we report on frequency doubling results by Cerenkov radiation from crosslinked polymer slab waveguides. The absorption by the

NLO polymer used for doubling of an Nd-YAG laser is small even at the SH wavelength ($\lambda = 532$ nm). For SHG of a 830-nm diode laser, a polymer with a larger NLO coefficient was used. The green and blue light were observed and the conversion efficiencies were found to be relatively high.

2. Sample preparation

Figure 1 shows the structures of the NLO polymers used. They are epoxies of Diglycidyl Ether of Bisphenol A and 4-nitroaniline or 4(4'-nitrophenylazo) phenylamine (Disperse Orange 3) functionalized with cinnamoyl groups (DGEBA-NAC or DGEBA-DO3C). Photocrosslinking is realized by 2+2 addition through the cinnamoyl groups. The detailed synthesis of these polymers is reported elsewhere.[8] The number density of NLO chromophores in DGEBA-NAC has been increased compared to the materials reported earlier.[9] Figs. 2 and 3 show absorption spectra of DGEBA-NAC in tetrahydrofuran (THF) and a DGEBA-DO3C thin film, respectively. The peaks at 278 nm are due to the crosslinking groups. Figure 3 also shows the spectrum of the film irradiated for 10 min by a 3 mW/cm^2 UV light with emission peak at 254 nm.

figs. 1-3

table I

Films were prepared on 1-mm-thick end-polished glass substrates by spin coating a solution of a polymer in propylene glycol methyl ether acetate (PGMEA) (weight ratio 1:5.5 and spin speed 4000 rpm for DGEBA-NAC). The samples were placed in a vacuum oven at 40 °C for 12 hours to remove

residual solvent. Glass transition temperature T_g of the polymers in the uncrosslinked state was measured by a differential scanning calorimeter. Refractive indices n of the polymer and the substrate were measured by an ellipsometer and are summarized in Table I along with the other results. The film thickness was estimated to be 0.5 μm from the interference pattern in the transmission spectrum.

Samples were poled by corona discharge in wire-to-plane configuration and the detailed set-up has been reported earlier.[12] A tungsten wire of 100-μm diameter at a high potential (~ 5 kV) was positioned above the polymer placed on a hot plate. The sample was slowly heated (~ 15 min) from room temperature to the poling temperature which was 5 °C lower than T_g. After 5 min of poling, crosslinking was performed by UV irradiation with the poling field turned on. The light source was a mercury lamp producing an intensity of 1 mW/cm^2 on the polymer with emission peak at 254 nm. An exposure time of 5 min was determined to be optimum for crosslinking and limiting the photodegradation of the NLO chromophores. The sample was cooled down to room temperature in about 5 min. NLO properties of poled films have been measured by Maker fringe SHG. The detailed experimental arrangement and calculations of the NLO coefficient d have been described elsewhere.[12-14] The measured d_{33} values are also given in table I.

3. Cerenkov SHG

Fig. 4 schematically shows the experimental arrangement of frequency doubling. A waveguide is formed by three media: substrate, polymer, and air. Since d_{33} is the largest SHG tensor element of a poled film, the TM_0 waveguide mode was excited through a polarizer. A lens with a focal length of 333 mm was used to focus the beam weakly and a rutile prism was utilized to couple light into the waveguide. The waveguide streak was monitored using a CCD camera. The condition for the guided modes of the fundamental light and for Cerenkov SHG is expressed as

$$n_s(\omega) < N(\omega) = n_s(2\omega)\cos\phi < n_s(2\omega) \quad (1)$$

where n_s is the refractive index of the substrate, N is the effective index, and ϕ is the angle between the waveguide and the SH beam in the substrate. By calculating the TM_0 dispersion equation, it was found that the above condition is satisfied if a DGEBA-NAC film thickness is between 0.36 and 0.55 μm at a fundamental wavelength of 1.064 μm.

figs. 4-6

Frequency doubling of 1.064-μm light using DGEBA-NAC was carried out. The light source was a Q-switched Nd-YAG laser, with a pulse width of 200 ns and a repetition rate of 50 kHz. When measuring the SH power, the fundamental wave was blocked by filters. Fig. 5 shows the measured behavior of the SH power as a function of the fundamental power on logarithmic scales. The slope of the data points confirms that the SH power scales as the square of the fundamental power. The SH peak output corrected for the filter loss was 470 nW at a fundamental peak input of 18.6 W incident on the prism. The total conversion efficiency normalized by the

fundamental power was 1.7×10^{-7} %/W. The fundamental peak output at the waveguide edge was 0.032 W. Thus the waveguide conversion efficiency is calculated to be 5.6×10^{-3} %/W, considering the measured waveguide loss of 2.4 dB/cm and an interaction length of 1.9 cm for the fundamental wave. These efficiencies are significantly larger than those reported earlier,[4] mainly due to the longer interaction length and the lower SH absorption by the NLO polymer used in the present work. Fig. 6 is a photograph of the prism-waveguide clamp, where the frequency doubler is emitting green light from the substrate edge. Green light was observed even when the laser was operating at a continuous wave mode.

fig. 7

To obtain blue light, a continuous wave 830-nm diode laser was used as the light source. Since the laser power is much smaller, DGEBA-DO3C was selected to fabricate the frequency doubler because of its larger NLO coefficient. The SH output was 28 pW at a fundamental input of 10 mW. The total and net waveguide conversion efficiencies were 2.8×10^{-5} %/W and 0.03 %/W, respectively. Fig. 7 is a photograph of the blue light emitted from the doubler through a filter stopping the fundamental wave.

4. Conclusion

Efficient Cerenkov SHG of a 1064-nm Nd-YAG laser and a 830-nm diode laser was carried out in crosslinked poled polymer slab waveguides. For the YAG laser, the SH wavelength is away from the strong absorption region of the polymer used. Green light was observed even when the laser

was operating at a continuous wave mode. Blue light was obtained using the diode laser and a polymer with a larger NLO coefficient. The waveguide conversion efficiency was about 0.03 %/W with a total efficiency of 2.8×10^{-5} %/W. Future work includes making channel waveguides for improving the efficiency further.

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Figure captions

Fig. 1. Structures of the NLO polymers.

Fig. 2. Absorption spectrum of DGEBA-NAC in THF.

Fig. 3. Absorption spectra of a DGEBA-DO3C film before and after UV irradiation.

Fig. 4. Cerenkov SHG by NLO waveguide.

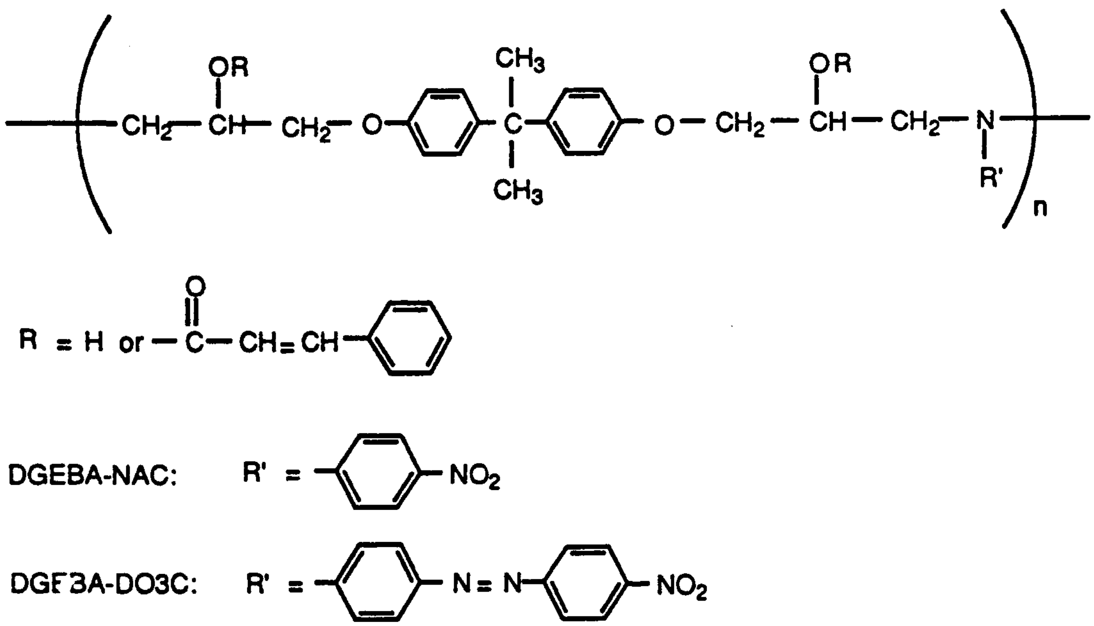
Fig. 5. SH power vs fundamental power on logarithmic scales.

Fig. 6. Photograph of the frequency doubler that is emitting green light.

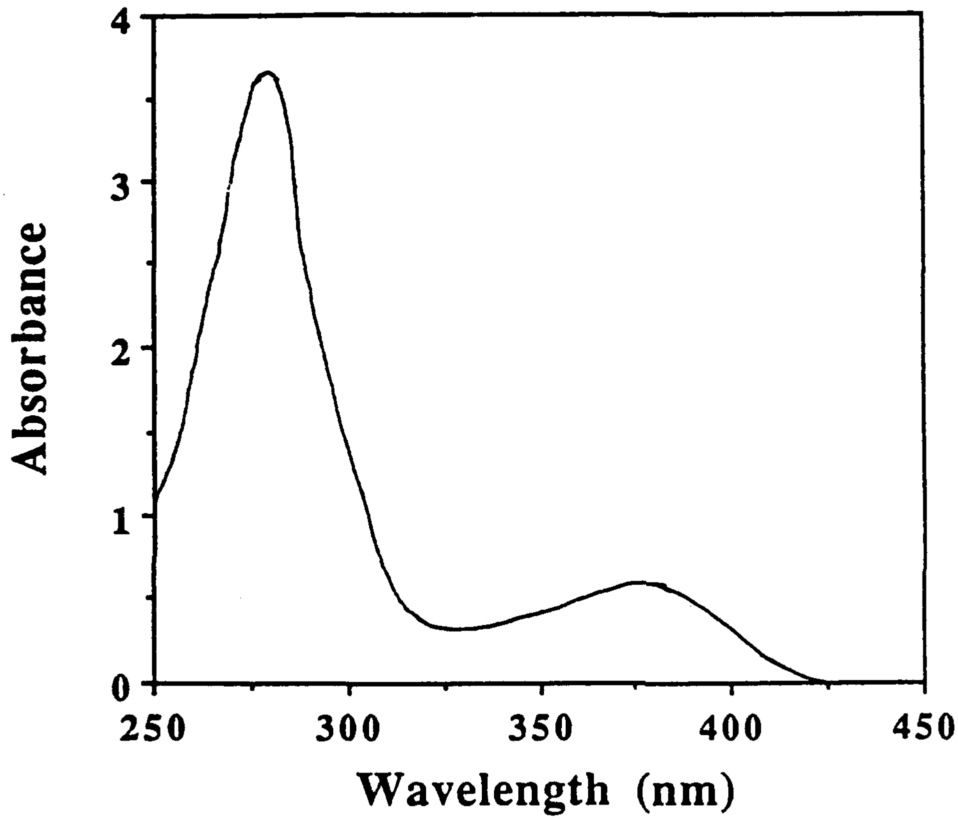
Fig. 7. Photograph of the blue light emitted from the doubler.

Table I. Properties of the NLO polymers and the substrate.

		DGEBA-NAC	DGEBA-DO3C	Glass
T_g (°C)		83	103	
n at $\lambda =$	415 (nm)			1.537
	532	1.637	1.718	1.522
	633	1.635	1.693	1.514
	830	1.618	1.658	1.505
	1000	1.613	1.652	1.502
Waveguide loss (dB/cm)		2.4 (λ =1064 nm)	1.6 (λ =830 nm)	
d_{33} (pm/V) (λ =1064 nm)		8.1	31	



X. Zhu et al., Fig. 1



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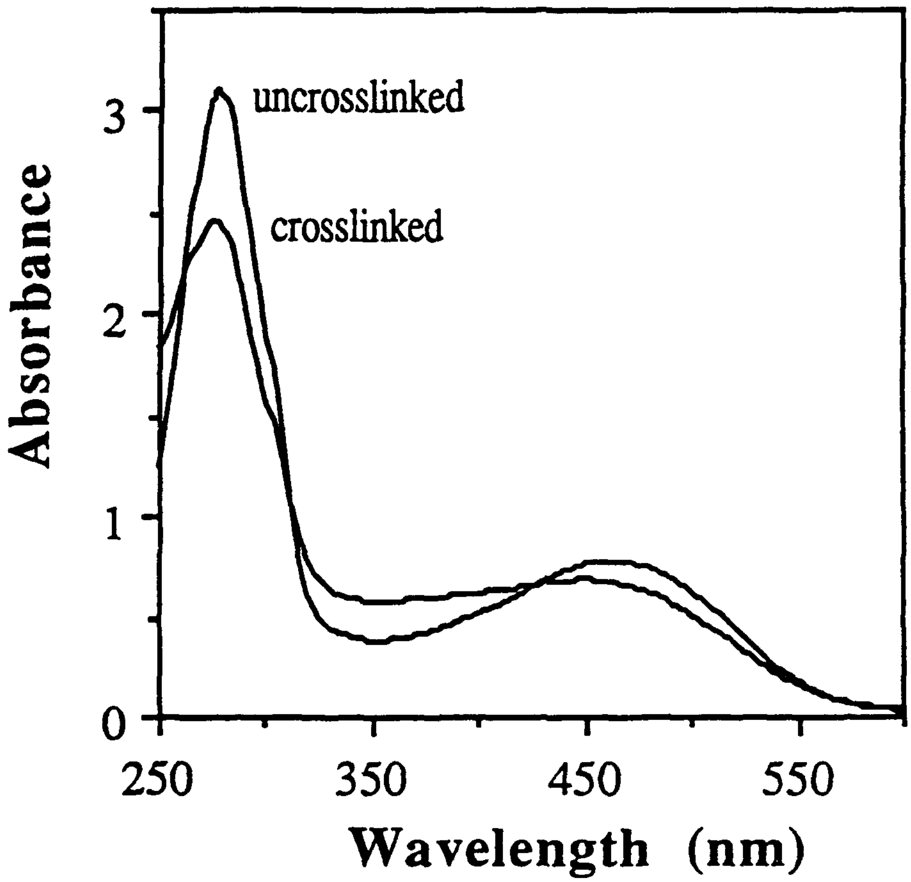

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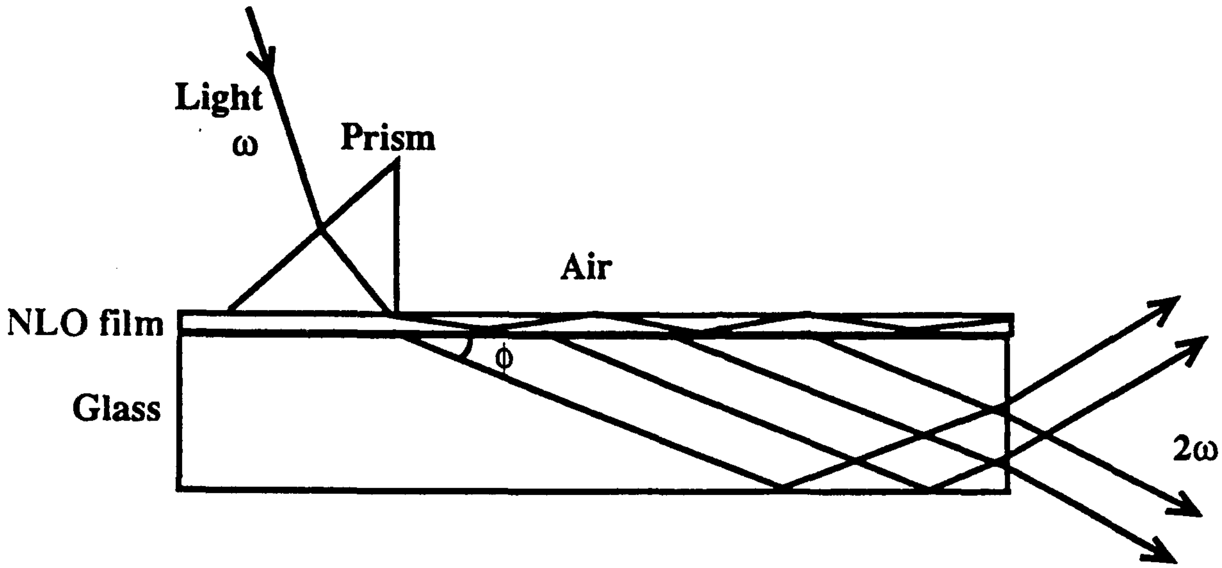

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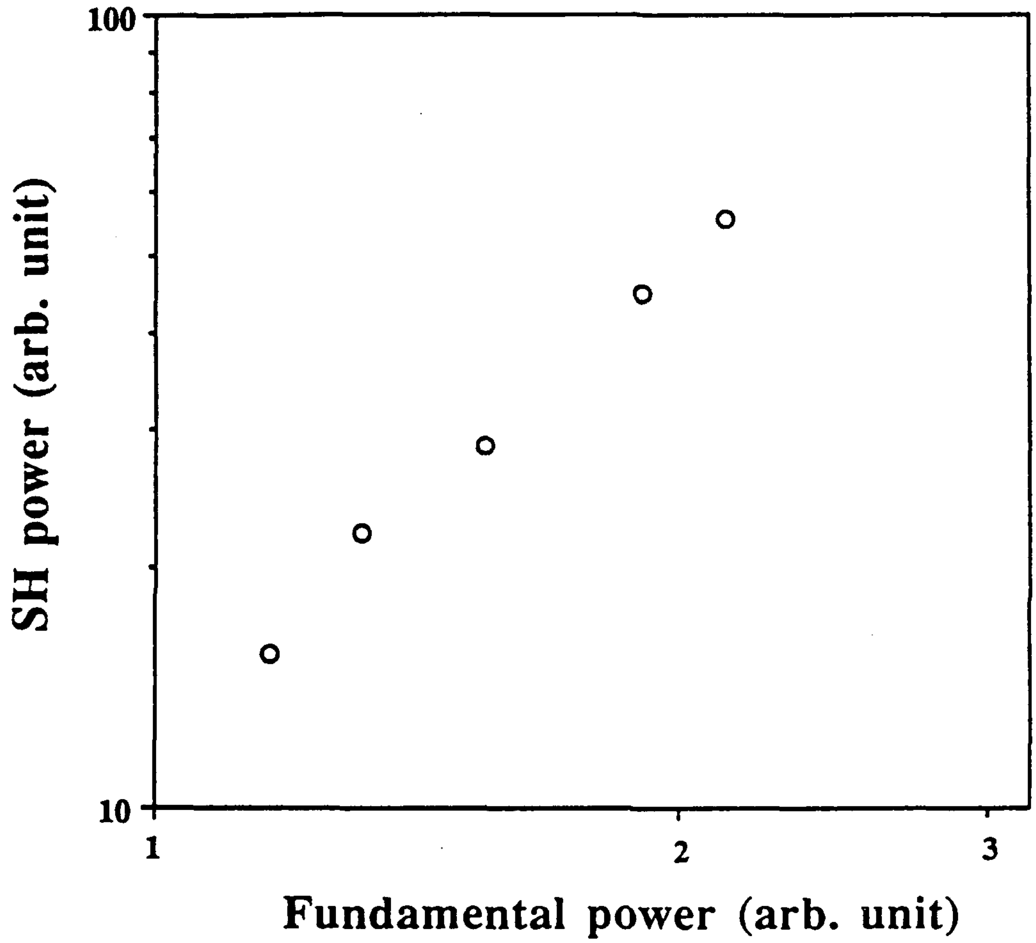

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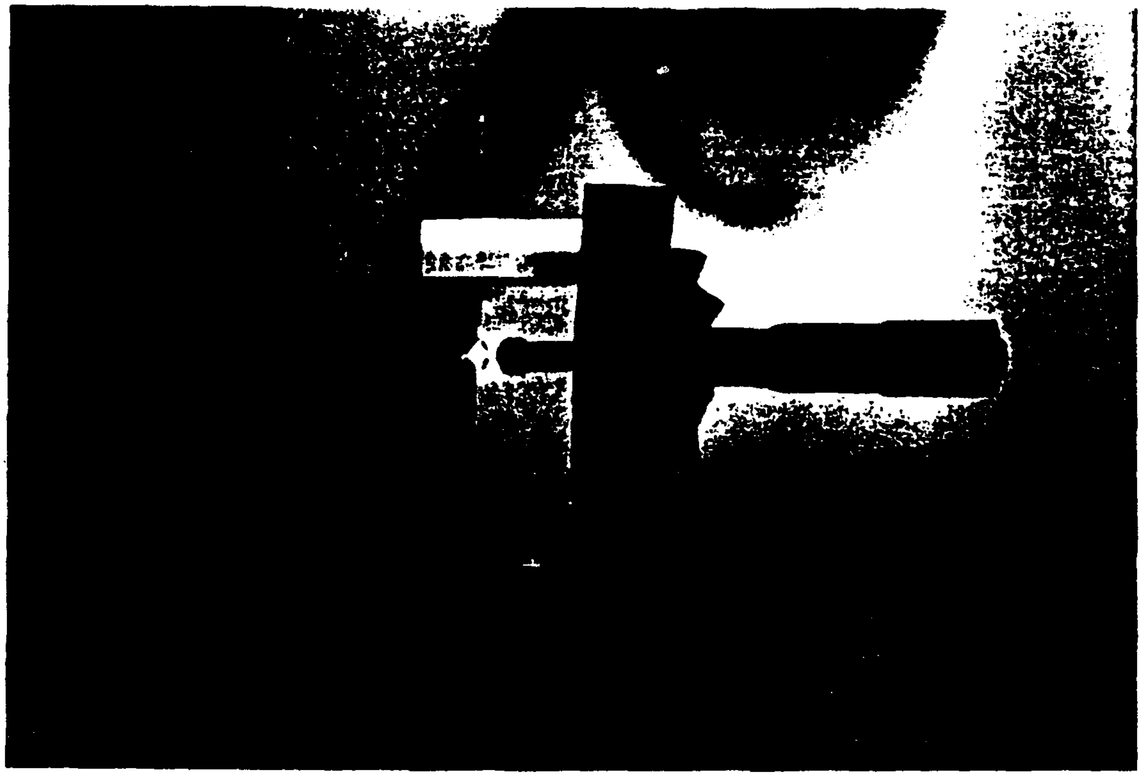

X. Zhu et al., Fig. 6

Fig. 7

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